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CO_2 sequestration with limited sealing capability: A new injection and storage strategy in the Pearl River Mouth Basin (China)

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ABSTRACT

Storing CO_2 for geological timescales is crucial to delivering decarbonisation of industrial economies aligned with global climate change mitigation objectives. This requires CO_2 injection into a large subsurface pore space overlain by a high capillary entry pressure seal rock to prevent CO_2 escape. Such formations occur abundantly in productive hydrocarbon basins such as the North Sea. However, these geological conditions for CO_2 storage are unavailable close to some large CO_2 emitting economic areas in China. We identify this problem and innovate a solution for the example of the Pearl River Mouth Basin (China). This offshore basin has an estimated effective storage capacity of hundreds of Gt CO_2 based on reservoir pore volumes. Production and geological data of three oil fields of the Huizhou hydrocarbon cluster are analysed as analogues for large-scale aquifer storage of CO_2. Results suggest the injection and storage of CO_2 in individual trap structures will perform poorly, due to thin reservoir layers, small structural traps, and indications that the cap rocks may have only limited sealing capability. To address this a novel alternative CO_2 sequestration strategy, based on multi reservoir layer injection and dispersion trapping, is recommended to exploit the vast available pore space for CO_2 storage.

1. Introduction

The 2015 Paris Agreement targets reduction of anthropogenic greenhouse gas emissions to hold the increase in the global mean temperature to below 2 °C above pre-industrial levels (United Nations, 2015). China became, and continues to be, the world's largest emitter of CO_2 in 2007 (The Netherlands Environmental Assessment Agency, 2007), mainly because of the country's dependence on coal as a source of energy with more than 900 GW of installed coal-fired power plant capacity and nearly 200 GW under construction (IEA, 2016). Carbon Capture and Storage (CCS) is widely considered a crucial technology to reduce CO_2 emissions in China from coal-fired power plants and other industrial sources and help China to meet its Nationally Determined Contribution commitment to the UNFCCC Paris Agreement (UNFCCC, 2017; Scott et al., 2013). The presence of saline aquifers as large-scale CO_2 storage sites within reasonable distance to major CO_2 emission clusters is key to enabling CCS delivery in China.

The Guangdong province in the south-east of China, with approximately 110 million citizens, is one of China's centres of industrial activity and associated emissions, making it a target region for the application of CCS on large point sources. The Haifeng full-chain offshore CCUS (Carbon Capture, Utilisation and Storage) Project is planned as an integrated CCS demonstration project to initiate CCS deployment in the region. This project involves the capturing of CO_2 from a combustion power plant located in Shanwei, transport of the CO_2 via a pipeline and CO_2 injection and storage in the offshore Pearl River Mouth Basin (PRMB) (Li et al., 2015). A preliminary assessment of the CO_2 effective storage capacity of the PRMB estimated it to be 308 Gt (P50) mainly in deep saline aquifers, with P15 and P85 probabilities of 473 Gt and 118 Gt of CO_2 respectively (Zhou et al., 2011). These very high capacity numbers are based on a volumetric approach typical for 'effective capacity estimations' using storage efficiency factors of 0.01, 0.026 and 0.04 for low-, mid- and high-case, respectively, taken from studies referenced (Zhou et al., 2011). These storage efficiency factors were not specifically calculated based on geological and geophysical data from this particular basin (Heinemann et al., 2017a). A more refined storage capacity estimation remains difficult because geological data is mostly owned by the state owned petroleum companies of China and is not freely available. Hence, the PRMB's very large CO_2 storage capacity estimation is based on limited data.

In this study we investigate the proximal part of the PRMB as a large scale CO_2 storage province based on the analysis of three oil fields of which two have been shortlisted as suitable targets suggested for CO_2 storage (Li et al., 2015). Our analysis is mainly based on available

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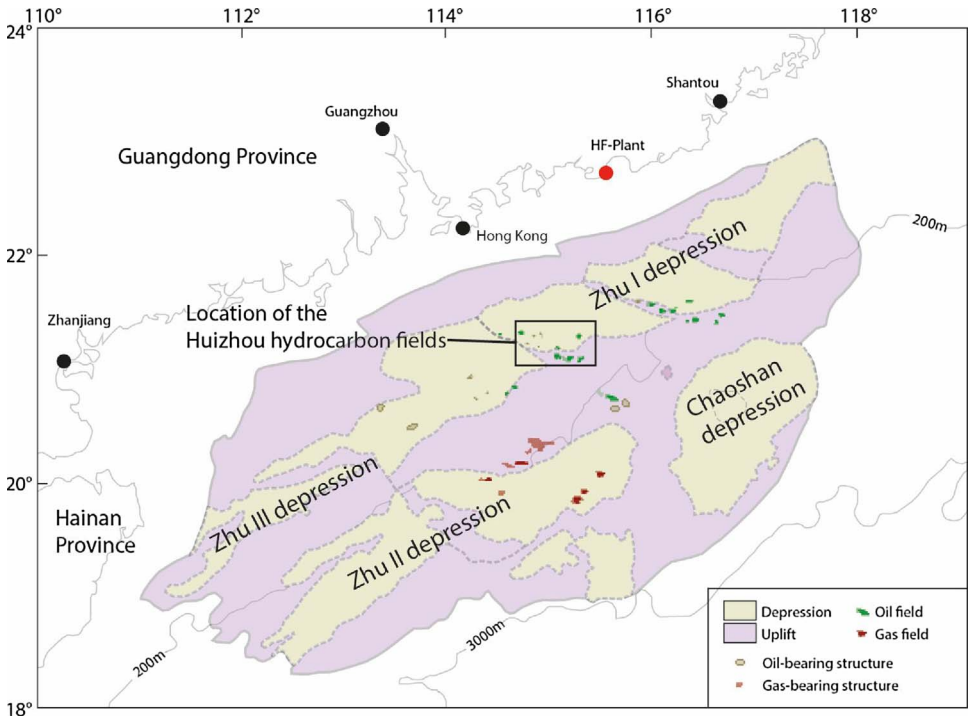

Fig. 1. Simplified geological map of the Pearl River Mouth Basin showing the location of the Huizhou hydrocarbon fields and the Haifeng Power Plant (HF-Plant). Modified after Li et al. (2015).

production data published in Chinese. We identify and discuss major geological obstacles which make secure CO_2 sequestration conditional, and are likely to prevent CO_2 storage as conventionally envisioned. For the PRMB, these are the lack of trap structures, insufficient thickness of individual reservoir formations and the presence of cap rocks with only limited sealing capability. To address these shortcomings, we propose and assess novel sequestration strategies to unlock the, certainly available, vast pore space of the PRMB for geological storage.

2. The Pearl River Mouth Basin

The PRMB is located in the northern South China Sea offshore Guangdong close to major cities such as Hong Kong and Shenzhen. It is an approximately 900 km long and 115–280 km wide, ENE-elongated basin with a total area of roughly 175,000 km^2. It is a marginal rift basin that developed during Cenozoic times on a basement of Mesozoic granites and Palaeozoic metamorphic rocks (Zhou et al., 2011). The water depth ranges from 50 m to 2000 m, covering sediments of up to 6 km in the shelf and up to 14 km in the slope (Li et al., 2015; Zhou et al., 2009; Chen et al., 2003). The basin consists of two major depressions aligned within three ENE running uplift (Fig. 1) zones (Jiang et al., 2015). The geological evolution can be divided into a Paleogene rifting stage with mainly continental deposition in graben and half-graben structures when the source rocks of the Wenchang and Enping formations were deposited; and a Neogene thermal subsidence with widespread marine deposition (Dongdong et al., 2009). In reservoirs of the Zhu I depression, hydrocarbon expulsion and the migration of hydrocarbons from the source rocks along active faults to shallower Zhuhai and Zhujiang reservoir formations peaked at the end of the middle Miocene (Du et al., 2009). Horizontal secondary migration was likely to have occurred along permeable Zhujiang formations, as research on the Zhu III sub-basin indicates (Fengjun et al., 2001).

The hydrocarbon resources of the PRMB have been assessed to be 2.2 billion tonnes of oil and 743 billion m^3 of gas. The proven oil reserves are 0.58 billion tonnes of oil and proven gas reserves are 58.6 billion m^3 according to a survey conducted in 2008 (MLRC, 2008). Exploration started in 1990 and was focused on proximal shallow water plays, but attention switched to the deep-water after the discovery of the Liwan gas field in 2006. This discovery, located 350 km southeast of Hong Kong at a water depth of 1300 m was a play opener with a possible contingent resource of four to six trillion cubic feet of gas. The only significant oil discovery to date is the Liuhua 11-1, a field with proven reserves of over 155 Mt in a large structural closure within the carbonate platform located in the Western part of the Dongsha Massif, 220 km southeast of Hong Kong (Tyrrell and Christian, 1992). All other oil discoveries in and around the Zhu I depression are smaller (Zhu et al., 2007). A published list shows that 9 out of 50 oil fields have oil reserves of more than 17.2 million tonnes (20,000*10^3 m^3) each in the Zhu I depression (Zhu et al., 2007). If, as an assumption, 40% of the 17.2 million tonnes of oil of an oil field can be produced and replaced with CO_2, less than 7 million tonnes of CO_2 can be stored in an oil field considered in the top 20% range in terms of the oil field size. Hence, storage potential in depleted hydrocarbon fields close to the Haifeng Power Plant and potential follow-on commercial scale CCS projects is limited (Heinemann et al., 2017b). More recently, large gas fields have been discovered in deeper water (Zhou et al., 2011) which suggests that exploration and development of the basin has not reached a mature stage. For the Zhu III depression, it is assumed likely that a large amount of hydrocarbon has either been lost or is to be found in undrilled structures (Fengjun et al., 2001).

The apparent lack of large oil and gas accumulations in the proximal part of the PRMB and the determining factors controlling the small size of discovered hydrocarbon accumulations are important for understanding the feasibility of CO_2 sequestration. Here, we analyse three oil fields (HZ21-1, HZ32-2 and HZ32-3) in the Huizhou hydrocarbon field cluster, one of the main targets for future CO_2 storage in the PRMB, to identify reasons for limited hydrocarbon accumulation and to assess the resulting implications for CO_2 storage in saline aquifers.

Firstly, the mostly deltaic and shelf marine reservoir formations consist mainly of thin sand layers. According to data taken from Liu (2011) all three investigated fields are within the Zhujiang Fm and the thickness of the oil field reservoir layers average 4.6 m, for the three analysed fields with a maximum of 9.2 m and a minimum of 0.9 m (Supporting information A). The stratigraphy of the PRMB is dominated by kilometres of alternating shale- and sand-rich layers with some of those sand-rich reservoir layers filled with hydrocarbons. Fig. 2 show

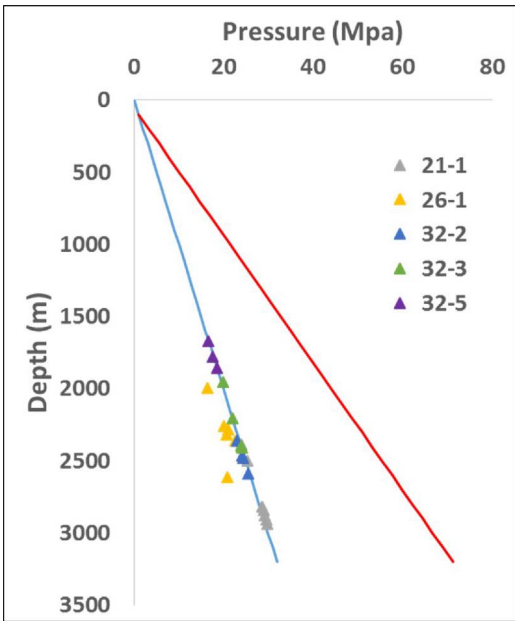

Fig. 2. Pressure data from the Huizhou oil fields. Most of the data plot on a hydrostatic pressure gradient (blue; calculated according to Liu (2011)). The data suggest that hydrostatic pressure gradient which suggests that no continuous hydrocarbon column between the oil layers exists. Also shown is a lithostatic pressure gradient (red; 0.023 MPa/m). Pressure data extracted from Liu (2011).

pressure data taken at virgin discovery pressures in the different reservoir layers lie on a hydrostatic pressure gradient which suggests that no continuous hydrocarbon column between the oil layers exists (Liu, 2011). The data also does not show significant reservoir overpressure, which is an indicator for an open system where injection induced overpressure can partially dissipate (Heinemann et al., 2012; Zhou et al., 2008). However, layer M20, the thickest of the initially oil filled layers of HZ32-2 with a thickness of 9.2 m, is significantly thinner than net reservoir thicknesses regarded as ideal for CO_2 injection and storage according to Chadwick et al. (2008).

Secondly, the anticlines in which most of the oil accumulated in the Huizhou hydrocarbon field cluster, have small structural closures. According to Liu (2011), the height of the structural closures average 22.1 m for the three fields with a maximum of 45 m and a minimum of 10.5 m (see supporting information A for detailed results). Structural closure data for HZ32-3 was not available and oil column heights were used instead; hence the average and maximum structural closure might be higher. Nevertheless, the structural closures are small compared to CO_2 storage targets such as the Goldeneye Field (Moray Firth, North Sea) with an initial gas column height of approximately 400 ft (122 m; Wilson et al., 2005).

3. Sustainable CO_2 column heights

Several reservoir layers of the analysed fields are not filled to spill and hence suggest capillary leakage of oil (Naylor et al., 2011; Fig. 3). No experimental analysis on core material was conducted to investigate threshold pressures of shales in the PRMB. Instead, sustainable CO_2 column heights are calculated based on oil field data from Liu (2011) (further detail on the methodology is in the supporting information B). CO_2 properties such as density and interfacial tension (IFT) were calculated using the Permedia CO_2 software package (http://www.permedia.ca/products/co2.html; Fig. 4) whereas the IFT of oil is assumed to be 0.03 N/m independent of depth.

It is assumed that a dynamic equilibrium exists between the capillary pressure (Pc), the pressure difference between the wetting and the non-wetting phase, and the threshold pressure of the cap rock (Pt). Hence any new oil migrating into the reservoir would cause capillary leakage to reach equilibrium. It is hypothesized, that the threshold pressure of all sealing layers of reservoirs which are filled to spill is in equilibrium with the capillary pressure. The sustainable CO_2 column height, a column height that does not cause capillary leakage of CO_2 into the overburden, is calculated as follows:

$$h_{CO2} = \frac{Pc_{oil} \cdot m}{\Delta\rho_{CO2} \cdot g}$$

Pc(oil) is the capillary pressure of the oil column at the reservoir-cap rock interface, m is the threshold pressure multiplier dependent on the IFT difference between oil and CO_2, $\Delta r(CO_2)$ is the density difference between CO_2 and brine and g is the gravity constant. The difference between the initial oil column height and the sustainable CO_2 column height is:

$$C = \frac{h_{CO2}}{h_{oil}}$$

According to this analysis, the sustainable CO_2 column heights of the fields HZ21-1, HZ32-2 and HZ32-3 will be 68%, 37% and 38%, respectively, of the length of the initial oil column heights. These values are average values for each field's individual layers. If the traps are filled up with CO_2 and the CO_2 column heights exceed the calculated sustainable column heights, capillary leakage will occur and CO_2 will migrate through the cap rock into the overlying strata. The parameters determining the change in column height from oil to CO_2 are the difference in density and IFT of the two fluids and the wettability. The higher the oil IFT in a brine dominated system, the lower the threshold pressure for CO_2 and hence the shorter the calculated sustainable CO_2 column height. The higher the oil density relative to the CO_2 density, the higher the threshold pressure for CO_2 and the longer the sustainable CO_2 column heights. The CO_2 system assumes a wetting angle of 37° imbedded in the Permedia software based on data from Chalbaud et al. (2009) and Chiquet (2006). The higher the CO_2 wetting angle, the lower the threshold pressure and the shorter the calculated sustainable CO_2 column height. The oil system is assumed to be water wet. Assumptions for the above calculation are detailed in the supporting information C.

4. Implication for saline aquifer storage

CO_2 sequestration is traditionally envisioned as the injection of large volumes of CO_2 into a reservoir formation overlain by a cap rock with a sufficiently high capillary entry pressure to retain the buoyant supercritical fluid (Haszeldine, 2009). This idea is based on the study and analysis of natural analogues mainly located in hydrocarbon rich basins such as the North Sea, the Norwegian margin and the oil rich American provinces. Some basins in the world which are close to large populous and industry centres are not large scale hydrocarbon provinces and have instead a geology which does not allow for this 'traditional' CO_2 sequestration concept. One example is south-western Australia where CO_2 storage with low capillary entry pressure cap rocks is currently under investigation (the South West Hub Project (Stalker et al., 2013)).

The Huizhou fields are recommended for CO_2 storage in the PRMB, mainly due to the moderate water depth (~100 m), location relatively close to CO_2 sources (~150–200 km to the Haifeng Power Plant) and the pre-existing oil and gas infrastructure. Two of the oil fields (HZ21-1 and HZ32-3) presented here have previously been shortlisted as suitable CO_2 storage sites for the first full-chain offshore Carbon Capture, Utilisation and Storage (CCUS) Project in Guangdong based on 'their potential of early abandonment, potential of CO_2 Enhance Oil Recovery application, suitability of geological conditions, and possibility for the reuse of oil and gas infrastructure for CO_2 injection and transportation (Li et al., 2015).

However, the analysis above suggests that CO_2 storage in the

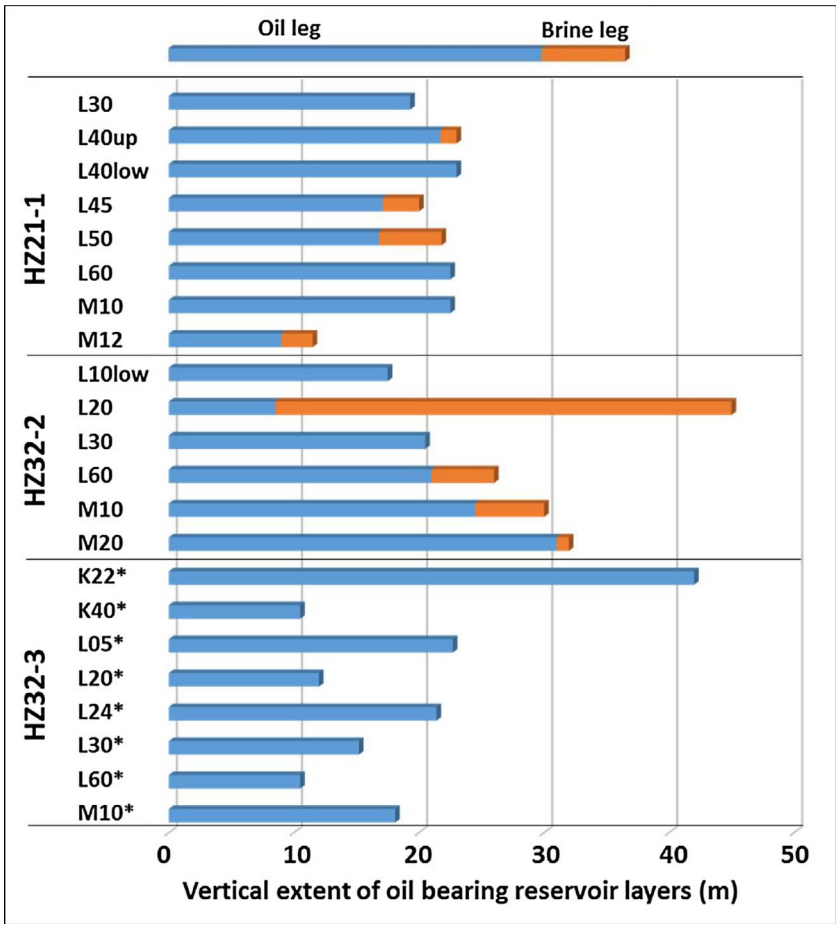

Fig. 3. More than half of the oil filled reservoir layers in HZ21-1 and HZ32-2 are not filled to spill. (*) No closure data for HZ32-3 is available and the vertical extent is assumed to be similar to the oil column height. Data taken from Liu (2011).

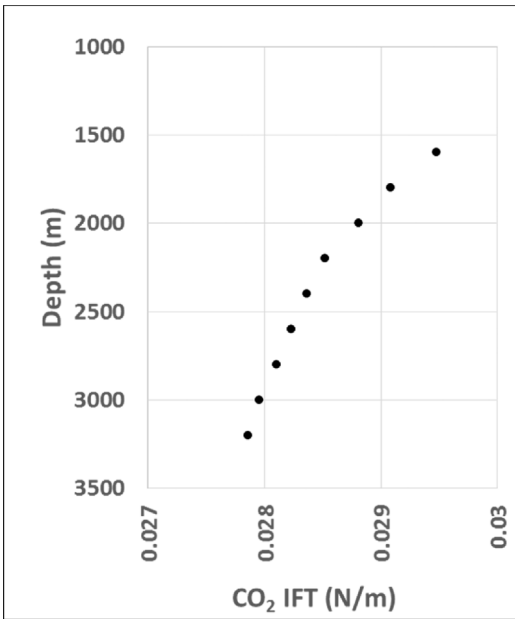

Fig. 4. Change of the interfacial tension of CO_2 and brine calculated by the Permedia software (Halliburton) for the geological conditions of the PRMB (see Supporting Information B for more information).

Huizhou area will have to take a distinctively different approach in design and planning to that applied to CO_2 storage in the North Sea such as the Sleipner and the Goldeneye project (Marshall et al., 2015; Michael et al., 2010). Firstly, in the fields investigated, there is no single extensive reservoir formation with a thickness that allows injection of millions of tonnes of CO_2 per year for a commercial CCS project with a multi-decade operating lifetime. Instead, the main oil bearing formations in the proximal part of the PRMB, the Zhuhai and the Zhujiang formations, are stacks of thin sand-rich and shale-rich sequences with thickness of the sand-rich parts less than a meter in places. The pressure data fall on hydrostatic pressure gradients and are interpreted as many thin unconnected hydrocarbon columns, with no single vertically-connected hydrocarbon column. Consequently, columns of stored CO_2 retained by these same seals will only be only a few metres in thickness. CO_2 injection into a single reservoir layer as it is done at the Sleipner injection site, where approximately a million tonnes of CO_2 per year are being injected into a vertically and laterally extensive high permeability sand reservoir, will not be feasible in the Huizhou area.

Secondly, oil has accumulated in anticlines with relatively small vertical closure of a few tens of metres. When combining the limited vertical closure with the relatively thin reservoir layers it becomes clear that the injection of CO_2 in single layers will very quickly lead to CO_2 migrating out of the trap structure such that control over the emplacement of the CO_2 will be lost.

Thirdly, and probably most importantly, our analysis shows that it is likely that the cap rocks of the three investigated oil fields are not capable of retaining long CO_2 columns (no calculated CO_2 column is longer than 20m). At locations where oil accumulations are present, the cap rocks could be oil wet and miscibility of injected CO_2 and oil could decrease the retention capability even further. To use them as large scale storage sites or even as injection points for aquifer storage will be both environmentally and economically risky. However, CO_2 storage in brine filled structures might also be challenging. The absence of hydrocarbons in a sedimentary basin can be due to several reasons including as hydrocarbon undersupply, charge timing and the absence of

traps. The frequent occurrence of only partially filled hydrocarbon fields, and the lack of large hydrocarbon fields, in the PRMB indicate that cap rock quality is generally poor in the proximal part of the PRMB and may be even poorer in areas with no hydrocarbon accumulations (Heinemann et al., 2017b). This may suggest that capillary leakage out of the storage site into the overburden is even more likely if CO_2 were to be injected into brine-filled structures. The results presented here cannot confirm the Upper Zhujiang Fm as an effective regional seal for CO_2 storage, as suggested by Zhou et al. (2011) and a detailed geological investigation in the retention capability of the second regional seal, the Upper Hanjiang Fm, is certainly required.

This analysis highlights the great uncertainties of basin wide "effective" CO_2 storage capacity estimations. These easy to conduct large scale volumetric approaches are extreme simplifications of complex and often uncertain subsurface geology. These simplifications will introduce uncertainties into calculations to investigate the future of energy and CO_2 emissions. It is therefore vital that basin wide CO_2 storage capacity estimations based merely on the volume of potential reservoir rocks or comparable parameters are used with great care as a more detailed geological analysis could alter the capacity significantly. According to our findings, the pore space in the PRMB will be more difficult to utilise for CO_2 storage than previously anticipated.

5. Alternative injection strategy

The known trap structures in the proximal part of the PRMB do not offer enough pore space to store large volumes of CO_2 given the assumptions about CO2 column height before cap rock breach. To utilise the substantial pore space available an injection strategy that maximises residual and dissolution trapping should be conducted.

Injection has to take place into several sand rich layers simultaneously in order to access multiple reservoirs, minimise near-bore pressure increase, and maximise dispersion of free phase CO_2 (Fig. 5). Similar to the secondary migration of oil in the Zhu I depression where oil spreads out into the sand rich reservoir layers (Jiang et al., 2015), CO_2 injected into deep reservoirs via deviated wells will migrate laterally into low amplitude traps driven by its higher buoyancy. The lateral migration of CO_2 along multiple thin low angle sand rich formations over 10 s of kilometres will enhance the dissolution of CO_2 into brine as well as enhance residual trapping in individual pores. The more the CO_2 migrates laterally and contacts the maximum amount of porewater, the more CO_2 can be stored. Since lateral migration is inevitable, a detailed investigation of the secure performance of potential vertical leakage pathways such as faults as well as sand injectites and igneous intrusions is necessary.

Additionally, vertical migration needs to be taken into account. The analysis of the three oil fields shows that capillary leakage is possible in reservoir layers where CO_2-brine capillary pressure exceeds the threshold pressure. This will allow CO_2 to migrate slowly into the overlying reservoir layers which might be only several metres above the initial accumulation. Once in the overlying layer, CO_2 will migrate laterally again leading to further dissolution and residual trapping. The main reservoir formations in the PRMB, Zhuhai and the Zhujiang formations, consist of hundreds to thousands of metres of thin sand and shale rich layers which, through this sequential method of lateral and vertical dispersion and associated dissolution and residual trapping, are able to store a significant amount of CO_2. This 'dispersion trapping', the storage of CO_2 during migration through a layered mixed- permeability formation with no effective extensive cap rock, could access the great potential of the PRMB for CO_2 storage.

A more detailed investigation of the geology, especially of possible vertical leakage structures such as faults and injectites, as well as of the cap rocks, is required before the full potential of the basin as a CO_2 sink is confirmed. For example, even where suitable cap rock exists studies have shown that faults and fractures may provide conduits for CO_2 leakage (Bond et al., 2017; Roberts et al., 2015; Miocic et al., 2014). A suitable analogue for future CO_2 sequestration projects in the proximal part of the PRMB will be the South West Hub Project in Australia. There, CO_2 is planned to be injected into a thick sequence of interbedded sandy and shaly sediments that will provide baffles to retain buoyant CO_2 and promote residual trapping (Flett et al., 2004; Hovorka et al., 2004). Similar to the PRMB, cap rocks are known to be thin but it is expected that the widely distributed interbedded shales and sandstones will retain injected CO_2 and allow safe, long term storage.

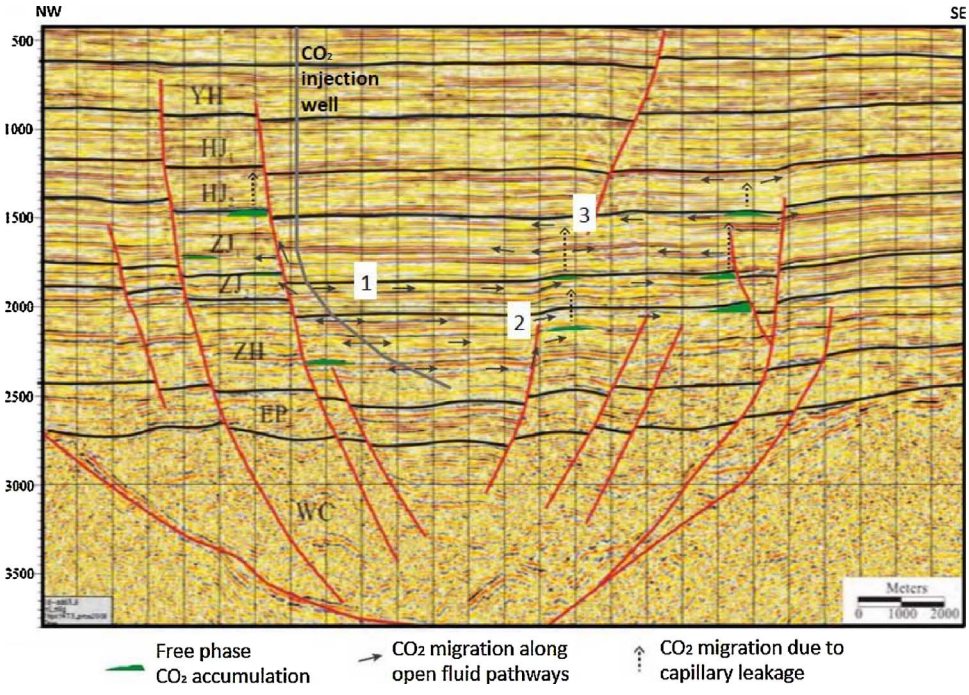

Fig. 5. The figure shows schematically the anticipated flow of a hypothetical CO_2 injection scenario in the proximal part of the PRMB and is for illustration purposes only. The original seismic section shows the Zhu 1 depression in the PRMB and was taken from Jiang et al. (2015 (Taylor & Francis Ltd, http://www.tandfonline.com)). CO_2 is injected at structurally deep parts of permeable formations such as the Zhuhai Fm and the Zhujiang Fm. (1) Near the injection well, CO_2 will predominantly spread out laterally along multiple thin reservoir layers. (2) Further away from the well, CO_2 will migrate up-dip along shallow angle reservoir layers over several kilometres. Faults may act as conduits to distribute into shallower reservoir layers or may block CO_2 flow depending on their permeability. (3) Whenever CO_2 accumulates in anticlines and other trap structures, the density difference between CO_2 and brine leads to rise in the capillary pressure. If the capillary pressure exceeds the threshold pressure of the overlying cap rock, capillary leakage will occur. Analysis in this article suggests that CO_2 column heights will be very small, typically 5–15 metres. CO_2 will migrate into the next overlying reservoir formation which might only be several metres above the initial CO_2 accumulation. The combination of lateral and vertical migration of CO_2 triggers an effective distribution of the injected CO_2 and will maximise trapping by capillary forces and enhanced dissolution of CO_2 into brine. The combination of CO_2 migration and structural, residual and dissolution trapping in heterogeneous formations has been named 'dispersion trapping'. CO_2 migration signs and accumulations are schematic and not to scale. WC – Wenchang Fm, EP – Enping Fm, ZH – Zhuhai Fm, ZJ – Zhujiang Fm, HJ – Hanjiang Fm, YH – Yuehai Fm.

6. Conclusions

- In three oil fields in the Huizhou area, candidates for CO_2 storage in the PRMB, pre-production pressure data imply that columns of stored CO_2 retained by the seals will be only a few metres in thickness, not many tens or hundreds of metres. Thus 'traditional' large-scale CO_2 sequestration considered in established North Sea or USA CO_2 reservoirs, where large volumes of buoyant CO_2 is injected in a thick reservoir formation and physically retained by high capillary entry pressures of a shale top seal, will not be technically feasible.
- The candidate storage sites in depleted oilfields are individually small and are suitable sites for a pilot injection projects; however much larger sites must be identified to host CO_2 from even a single coal power plant capture project. Reservoirs are thin, traps are small and cap rocks of limited sealing capability, and yet, cumulatively, massive pore space is available
- We suggest an alternative method of securely developing the resource of these sandstones for long duration CO_2 storage. The 'dispersion trapping' concept will enhance dissolution and residual trapping of CO_2 and will also enhance storage security and help to successfully access the vast pore space of the PRMB for CO_2 storage.
- The Australian South-West Hub injection experiments into poorly sealed reservoirs may provide useful analogues in the near future.

Acknowledgement

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.ijggc.2017.11.009.

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